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Spectral hole burning is one special holographic technology, which requires to cool down the holographic materia~ down to liquid helium temperature -270 degree of Celsius. However, it can record and generate very fast optical light pulse series and their interaction. Spectral hole burning is of critical interests for fast data storage and optical information processing. This work would be used to evaluate and study the general properties of the materials used for these application. This research is sponsored by the Air Force Office of Scientific Research under grant# F49620-99-1-0258.

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Large Scale Spectral Hole Burning Memories in Organic Materials

AFOSR Grant #F49620-99-1-0258

FINAL REPORT

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Contents

Theory – M# in Spectral Holeburning Material

We studied theoretically the M/# of multiplexing multiple holograms in the spectral hole burning material as absorption grating. According the bleaching curves got from the materials H_2-TBNP:PVB and Zn-TBNP:PVB, we proposed a theoretical model for the bleaching kinetics of the absorption bleaching. The amplitude absorption coefficient α decays exponentially as a function of the photonic energy exposure: $\alpha(r,t)=B+A\exp(-KI(r,t)t)$, where $I(r,t)$ is the light intensity pattern inside the material and 3K is a kinetic parameter of the material. For practical materials, there is non-bleachable absorption background B that persists after prolonged exposure.

For recording of a hologram pattern inside the material, the intensity pattern produced by the reference and signal beams R_i and S_I is $I(r,t)=I_I+V_I\cos(k_ir)$, where the modulation depth $V_I=2R_iS_i/(R_i^2+S_i^2)$. For simplicity, we ignore the non-uniformity of the intensity pattern due to the absorption inside the material, which is only a accurate approximation when the optical density is relatively small. For large optical density, the intensity pattern will not be uniform inside the material during the whole process of bleaching, and have to be simulated numerically, which we will investigate later.

With this theoretical approximation, we can derive the absorption grating strength for M equal holograms inside the materials as: $\alpha_M=B+\underline{\alpha}_0-\Sigma\alpha_i\cos(k_Ir)$, where the $\underline{\alpha}_0=A\exp(-KI_iM\tau)$ is the remaining absorption coefficient and the grating strength $\alpha_I=A\exp(-KI_IM\tau)\,KI_I\tau$. For transmission geometry, the diffraction efficiency is obtained using Kogelnick's expression: $\eta=\exp(-2Bd)\,\exp(-2\underline{\alpha}_0d)\,(\alpha_Id/2)^2$, where d is the normalized thickness of the recording medium $T/\cos\theta$, θ is the incident angle of the reference and signal beam inside the material.
M/# is normally used for measurement of the dynamic range of photorefractive materials used for multiple hologram recording, such as photo polymers and photorefractive crystals. We express the diffraction efficiency for absorption grating in similar expression as $\eta=(M\#/M)^2$, and derived the M# for the material. It turns out that M# is a constant, which only depends on the material parameter A,B and normalized thickness d. Therefore, M# is also a good dynamic measure for absorption material for multiple holograms recording, same as the other typical photorefractive materials for holographic recording. The M# for absorption material is derived as: $M\#=\exp(-Bd)AdC(Ad)/2$ where C(Ad) is a function of Ad given as:

$$C(Ad)=\max_{\beta}\left(\beta\exp(-\beta)\exp(-Ad\exp(-\beta))\right)$$

Figure 1 shows the simulation results for M# as a function of the optical density OD=2(A+B)d.

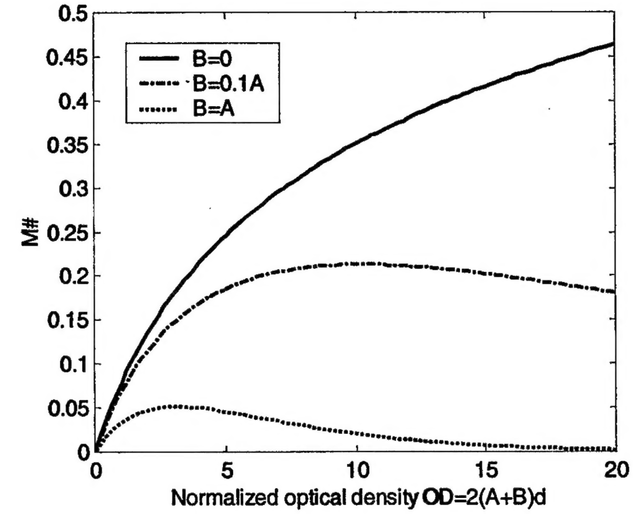

Figure 1. The M# as a function of the normalized optical density

To continue the theoretical studies of the M/# of multiplexing multiple holograms in the spectral hole burning material as absorption grating, we investigate the effects of the absorption during the recording process. Since the intensity pattern and the absorption coefficient inside the material are changing and effecting to each other during the bleaching process, we have to simulate the process numerically and compare with our previous simple theoretical model. With the same kinetic assumption for the bleaching property as: $\alpha(r,t) = B + A\exp\left(-K\int_0^t I(r,\tau)d\tau\right)$

where I(r,t) is the light intensity pattern inside the material and K is a kinetic parameter of the material. Considering the absorption on the recording beams inside the material, the intensity pattern for non-slanted transmission geometry is given as:

$$I(x,t) = I_0 \exp\left(-2Bd - 2\int_0^x \underline{\alpha}_0(l,t)dl\right)(1 + V_i\cos(k_i r))$$

during the recording of *i*th hologram. The modulation depth V_I is set to be 1 for all holograms. With this theoretical model, we can simulate the recording dynamics of multiple holograms inside the bleaching materials and calculate the M# as $M\# = M\sqrt{\eta}$

Figure 2 shows the simulated M# by equalizing 10 holograms inside absorption material as a function of the bleachable dynamic range OD_b=2Ad. Comparing with the results by previous theoretical approximation model also shown in the Fig. 2, the actual M# for B=0 is consistent with the theoretical model when OD<2, and then reaches a saturation value M#=0.2 at the optical density around 10. The detail simulation of the recording time and the grating strength inside the material show that the recording time for each hologram is similar when the optical density is small, and the grating strength is uniform inside the material, which matches the assumption of the model approximation. As the optical density getting larger, the extra part of the dynamic range is peeled off during recording of first hologram with extra longer time. All the gratings are finally located near a limited range near the end of the material, and all the other area was bleached uniformly without grating recorded.

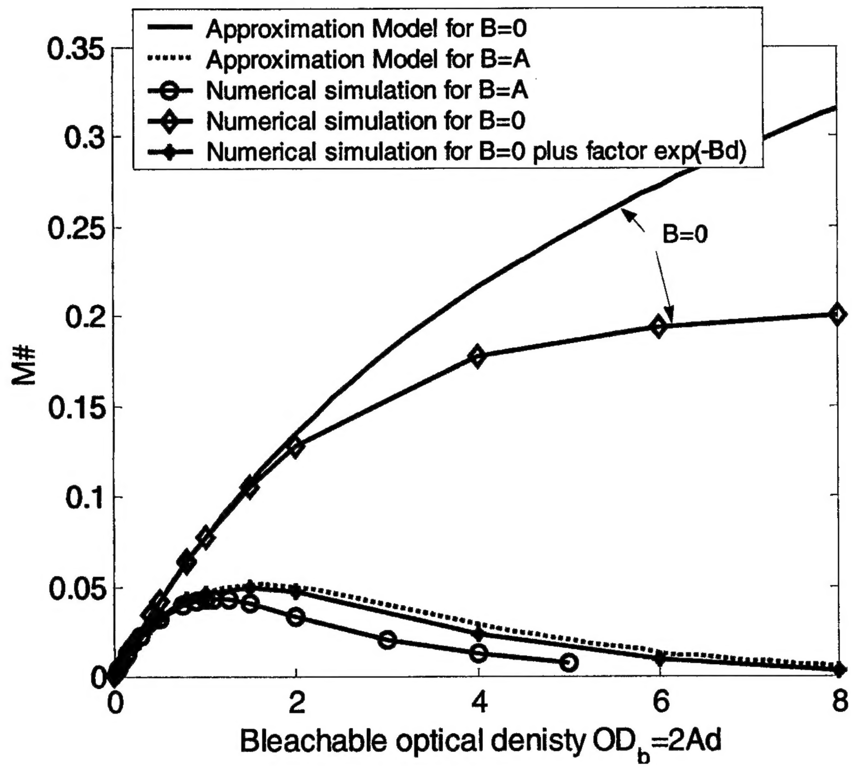

Figure 2. M# for absorption holograms

For B=A within the optical density, the M# is consistent with the theoretical approximation model as OD_b <1, and then getting smaller when OD_b getting larger. Simulation in Fig. 2 shows that the M# reaches a maximum 0.043 at optical density OD_b =1.1. It is smaller than the $M\#_{b=0}$ exp(-Bd) from theoretical approximation, because the actual non-uniform recording in the absorption material. The independence of the M# on the number of holograms M is also confirmed in the simulation of the dynamic recording. Fig.3 shows the M# for recording M=2, 5, 10 or 20 holograms in the absorption material with bleachable optical density OD_b=1.

For large-scale data storage application with spectral hole burning materials, it was demonstrated that the optimal method is to use frequency and phase swept (FPS) holograms. FPS use multiple holograms within nearby frequency range to enhance one hologram recording, with the advantages of larger diffraction efficiency, smaller cross-talk and less sensitive to readout erasure. The principle is due to the co-existence of an absorption grating and an index grating inside the material by the Kramers-Kronig dispersion relation. By adjusting the frequency and the phase relation between the multiple nearby holograms, they can superpose together and generate a strong phase grating at the central frequency, which is called π-phase shift holograms.

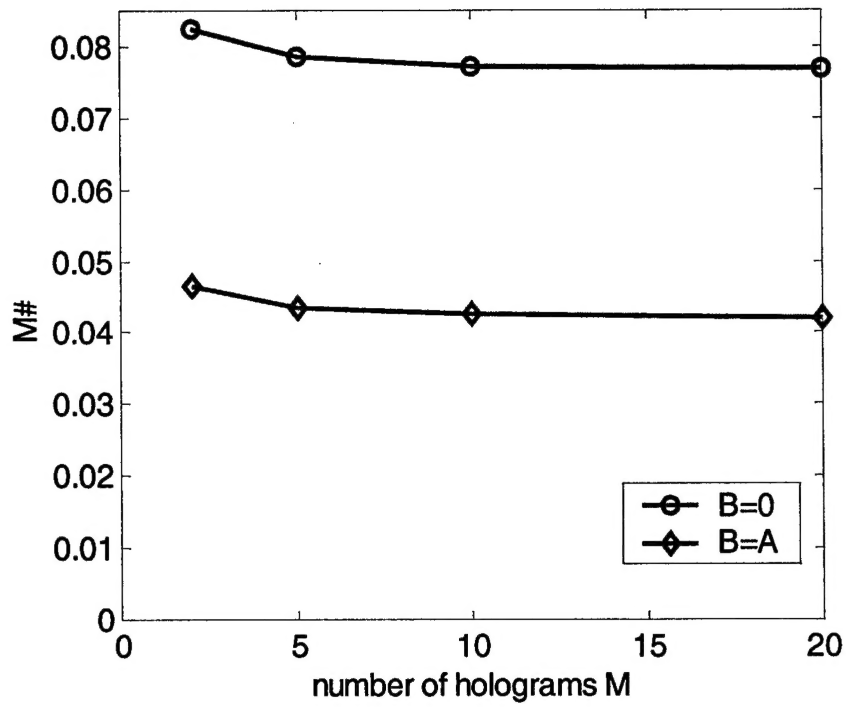

Figure 3. M# vs. the number of holograms multiplexed inside the material OD_b=1

Here we studies the simple two π-phase shift holograms to generate a single index grating hologram at the central frequency and calculate the diffraction comparing to the pure single absorption grating method. We start by assuming that the DC absorption $\underline{\alpha}_0$ and grating strength α_1 have a Lorentzian shape.

$$(A-\underline{\alpha}_0(\omega)) = A(1-\exp(-KI_i M\tau))\frac{\Gamma^2}{(\omega_b-\omega)^2+\Gamma^2}$$

$$\alpha_1(\omega) = A(1-\exp(-KI_i M\tau))KI_i\tau\frac{\Gamma^2}{(\omega_b-\omega)^2+\Gamma^2}$$

$$\beta_1(\omega) = j\alpha_1(\omega)\frac{\omega_b-\omega}{\Gamma}$$

where Γ is the homogeneous linewidth and ω_b the writing (burning) optical frequency. The index grating strength $\beta_1(\omega)$ is induced by the absorption grating by the Kramers-Kronig relation where ω is the read (probe) frequency. A Π-phase holograms recorded with a first exposure at frequency ω_1 and second exposure (same exposure energy) at frequency ω_2. When readout the hologram at the frequency $\omega=(\omega_{b1}+\omega_{b2})/2$, the absorption grating cancelled each other and the hologram is a pure phase grating as:

$$\beta(\omega) = j\alpha_1(\omega_{b1})\frac{2\Gamma\Delta\omega}{\Delta\omega^2+\Gamma^2}$$

where the $\Delta\omega=(\omega_{b2}-\omega_{b1})/2$,

The diffraction efficiency of this pure phase grating given by the Kogelnick's expression:

$$\eta = \exp(-2Bd)\exp(-2\underline{\alpha}_0 d)\left(\frac{\alpha_1 d}{2}\right)^2\left(\frac{2\Gamma\Delta\omega}{\Delta\omega^2+\Gamma^2}\right)^2.$$ The diffraction efficiency η has a maximum at $\Delta\omega=\Gamma$, and has the exact the same results as the single absorption grating.

However, one advantage of the FPS phase grating hologram is that the hologram is readout at the tail of the absorption grating, so it's less sensitive to the erasure during the readout. The optimal case is that the absorption can be totally bleached at the frequency where the hologram is readout without significant effects on the phase grating strength. The corresponding diffraction efficiency would be:

$$\eta = \exp(-2Bd)\left(\frac{\alpha_1 d}{2}\right)^2\left(\frac{2\Gamma\Delta\omega}{\Delta\omega^2+\Gamma^2}\right)^2.$$

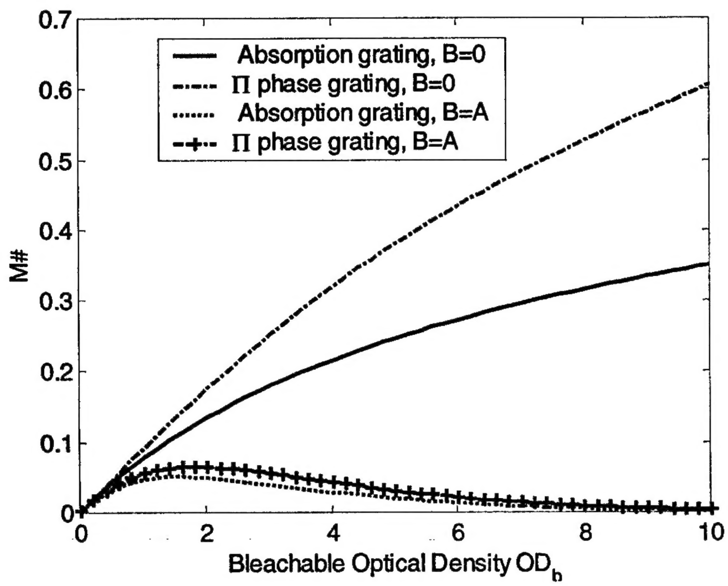

Figure 4. Comparison between the M# obtained when multiplexing Π holograms and a pure absorption holograms.

This expression is identical to the expression found for a pure absorption hologram, except for the absorption term in the exponential. If B=0, the medium is transparent and the diffraction efficiency is proportional to the optical density. Fig 4 shows a comparison between the M# obtained when multiplexing Π holograms and a pure absorption holograms. The M# of absorption holograms is lower than for index holograms indicating the advantage of multiplexing using Π holograms. However for the cases when there is a residual absorption not bleachable, the difference between the pure absorption grating and the Π phase grating is insignificant.

M# Measurement

From earlier work we know that the optimal bleaching intensity for H2-TBNP:PVB is about $10\mu w/cm^2$. We find that the optimal recording time lies between 5 seconds and 8 seconds where we get diffraction efficiency of about 10^{-4}. A Fourier lens is added behind the material in order to collect the reconstructed signal. In order to reduce the scattering noise we put an iris at the back focal plane of the lens. Fig.5 shows the selectivity with the iris. The selectivity is about 0.2 degrees. Fig. 6 shows the reading curve of the holograms. The diffraction efficiency gradually decays because the reference beam uniformly bleaches the material.

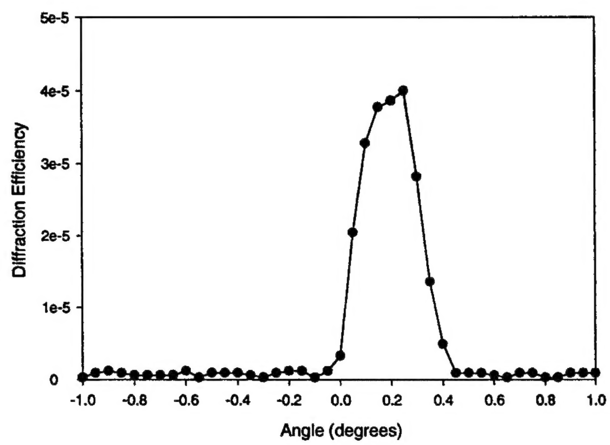

Fig. 5 Angle selectivity with iris

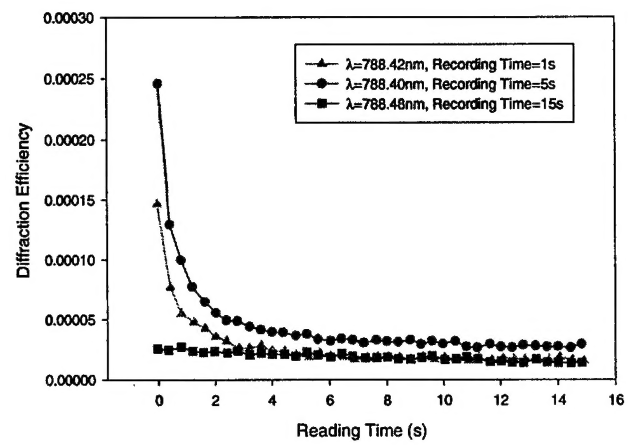

Fig. 6 Reading curve

The M/# of the spectral hole burning material was studied experimentally. From single hologram diffraction efficiency we estimate the M/# is about 0.01. Fig. 7 shows multiplexing of 10 holograms. The measured M/# is shown in Fig.8. The M/# has a trend to increase when multiplexing 3,5,7 and 10 holograms.

The measured M/# is about 0.01 which is also consistent with the theoretic calculation obtained from the bleaching kinetics of the material. Angle multiplexing can also be combined with frequency multiplexing to increase the storage capacity by a factor of 10.

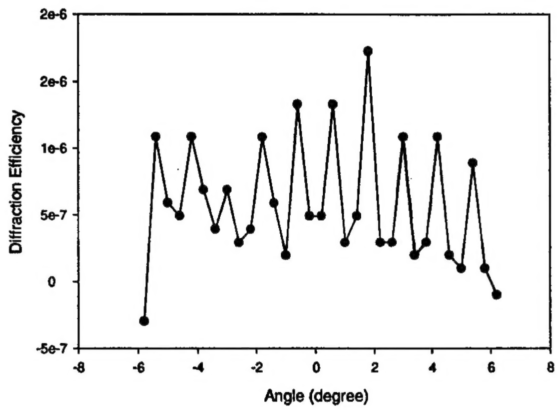

Fig. 7 Multiplexing 10 Holograms

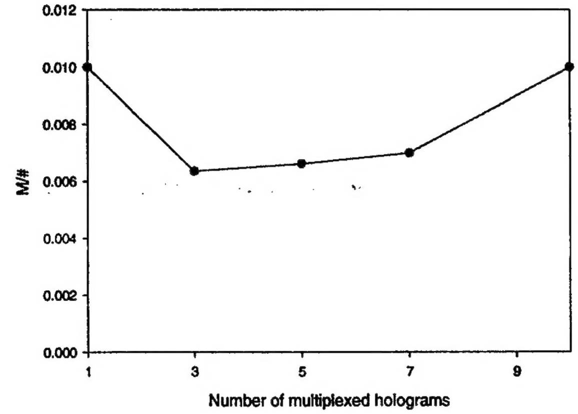

Fig. 8 M# experiment

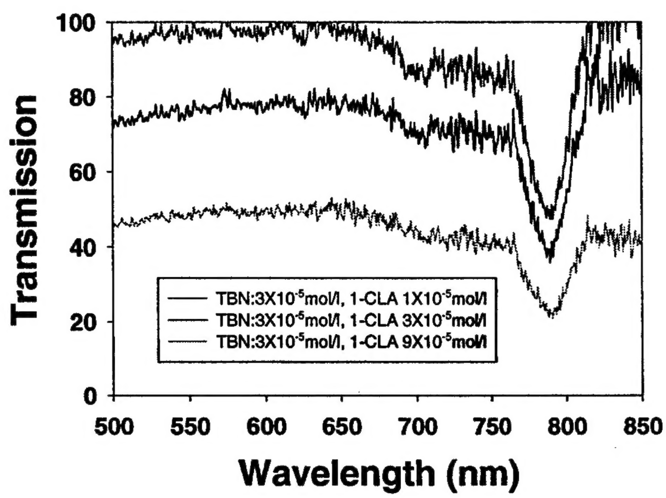

Figure 9. Transmission spectrum of 1CLA samples (PVB host)

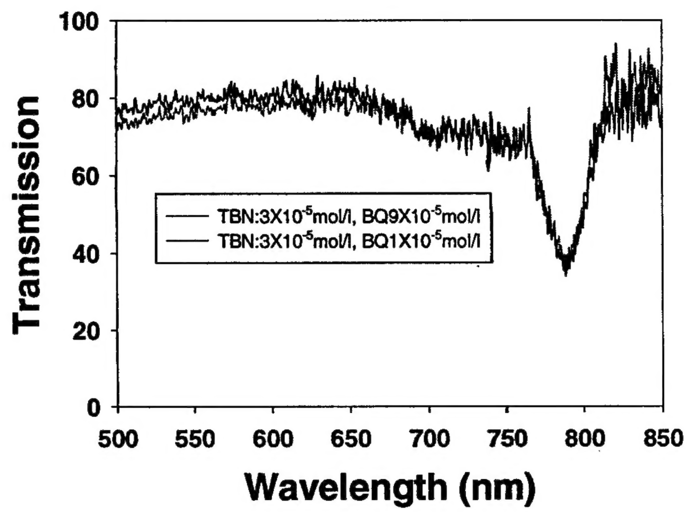

Figure 10. Transmission spectrum of BQ samples (PVB host)

Holograms recorded in spectral hole burning medium can be erased during readout when a uniform reference wave is illuminating the material. This is because the uniform reference beam continues to bleach the material. One idea to alleviate the problem is to develop a material which needs a separate sensitizing beam in order to record or erase holograms. During recording, the sensitizing beam, reference and signal beams are incident on the material and holograms can be recorded. Upon reconstruction, only the reference beam is incident on the material. Because the sensitizing beam is absent, holograms cannot get erased. We also continue to develop spectral hole burning material with high sensitivity. The following figures (Figures 9,10) show the transmission spectrum of our material under development.

Fast Movie Recording System

Since the measured M/# (0.01) of the spectral hole burning medium in one frequency channel is rather small and the readout process is destructive, we propose to use it in recording ultra-fast events instead of for memory applications. There are several important issues on recording femtosecond pulse holograms. First is the response speed of recording material. Second is the short coherent length of the pulse. Typically, we require the reference and signal pulses overlap with each other inside the recording medium in order to record their interference pattern or hologram. The hologram is only recorded in a thin slice and it reduces the multiplexing selectivity (either wavelength and angular selectivity). These problems can be solved by using spectral holography. Fig.11 shows our patent-pending optical design. A pulse from the regenerative amplifier is dispersed by a grating and focused by a lens. The back focal plane of the lens is the pulse spectral domain. We insert some delay line at the spectral domain (e.g. glasses with different thickness). A sequence of pulses are created and they do not have any common spectrum. We can use this sequence of pulses to record femtosecond movie. After recording, we use the oscillator to read out the hologram. The pulses from the oscillator go through the same pulse shaping setup and individual frame can be read out by blocking all but one of the delay lines. Spectral hole burning medium is a frequency selective medium. Holograms recorded by different spectral component of the pulse are stored in different types of molecules. Because the sequence of pulses do not have common spectrum, no two frames are recorded in the same type of molecule and can thus reconstructed separately. Fig.12 shows the calculated signal pulse trains with different delay lines.

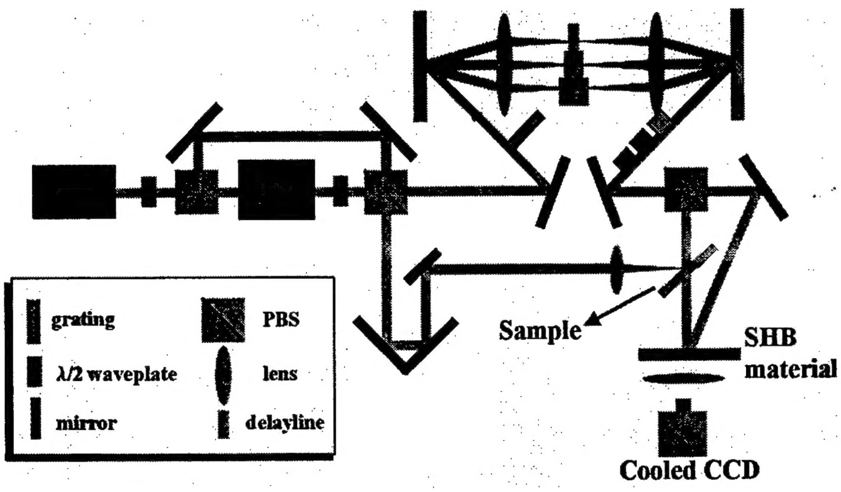

Figure 11. Fast holographic camera design.

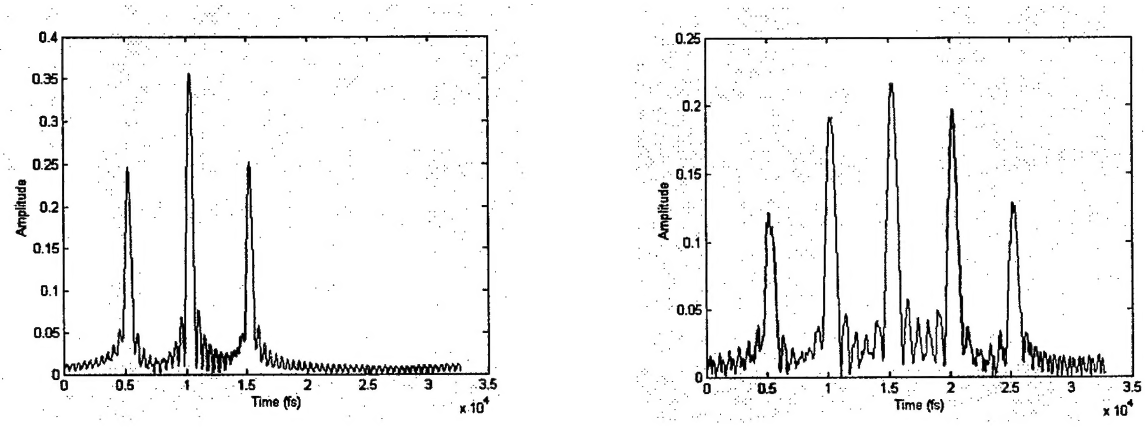

Figure 12. Calculated pulse train.

We generate the desired pulse train using the method we described in our previous report. The experimental setup is shown in Fig.13. A correlation setup is used to measure the pulse width and pulse seperation. Fig.14 shows a sequence of measured signal pulse train. The pulse widths of the pulses are larger than expected, possibly due to the dispersion of the lens. The pulse separation and amplitude are not uniform either. Future work will focus on the improvement of the system (replace the multiple glass plates with a mirror array, and use a true achromatic lens) and generating more pulses.

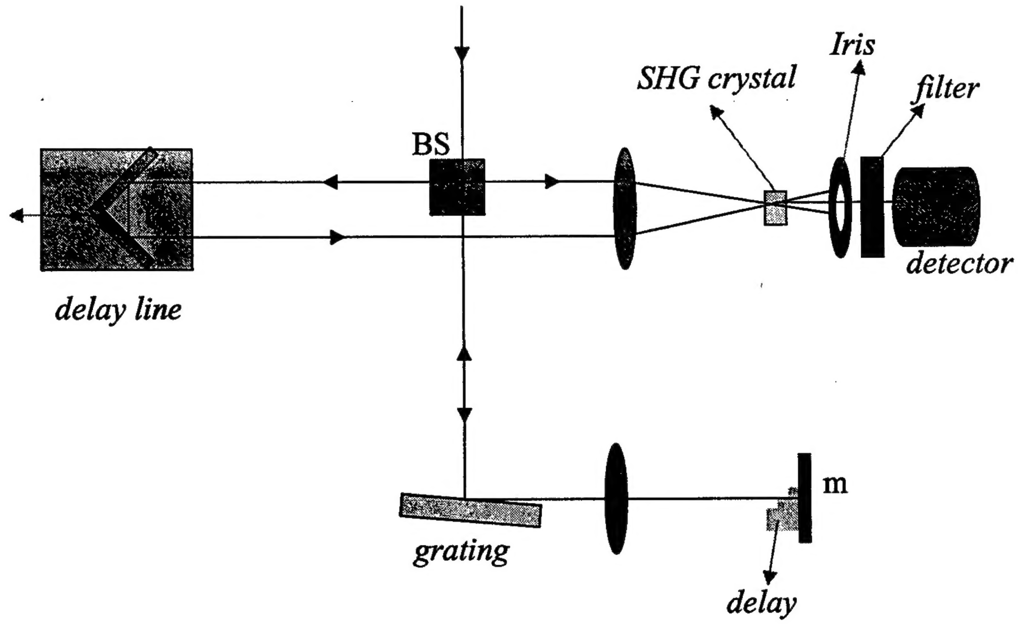

Figure 13. Setup for generating and measuring a pulse train

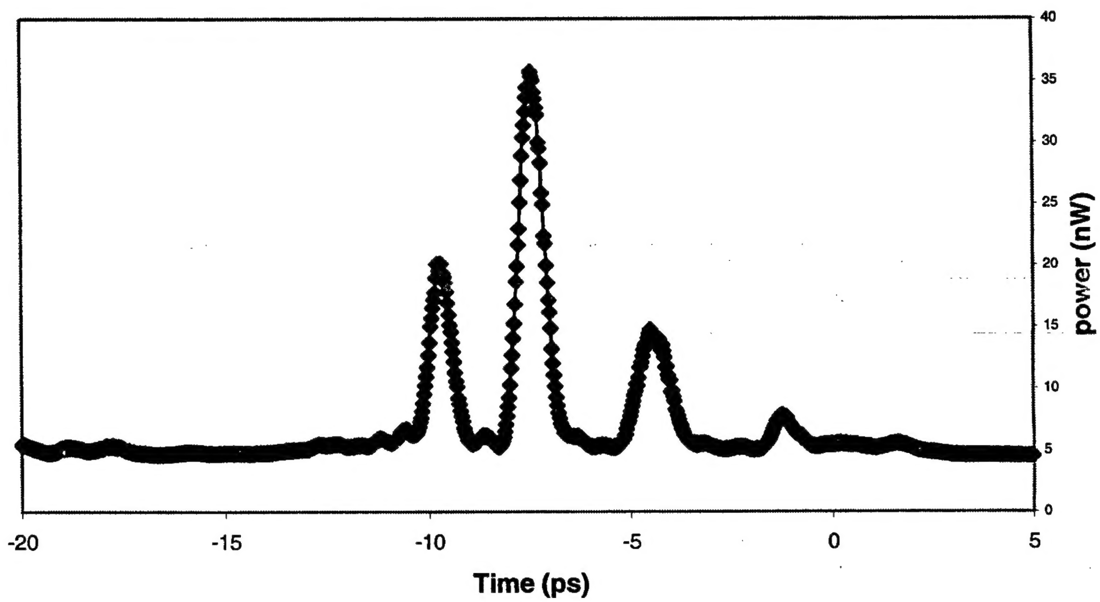

Figure 14. Measured pulse train profile

The pulse train generation mechanism is tested by recording three holograms of a stationary object on a CCD. The pulse train is generated using a grating and a mirror staircase, as shown in Figure15. After reflecting of the mirror staircase the pulses are

separated in a reference and signal branches using a beam splitter, and then recombined to record a hologram on a CCD (Figure 16). A transparency with an ellipse printed is used as the object (Figure 17). The reference beams are not plane waves since they do not go through the grating, and for this reason we can only recover a thin slice of the image from each hologram. Finally, we can digitally reconstruct the images from the recorded holograms (Figure 18).

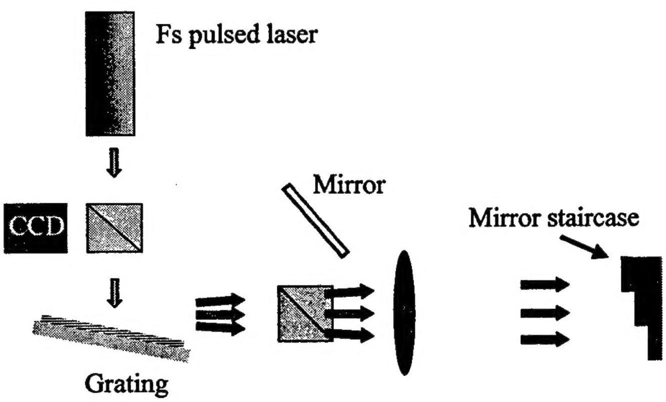

Figure 15. Pulse train generation.

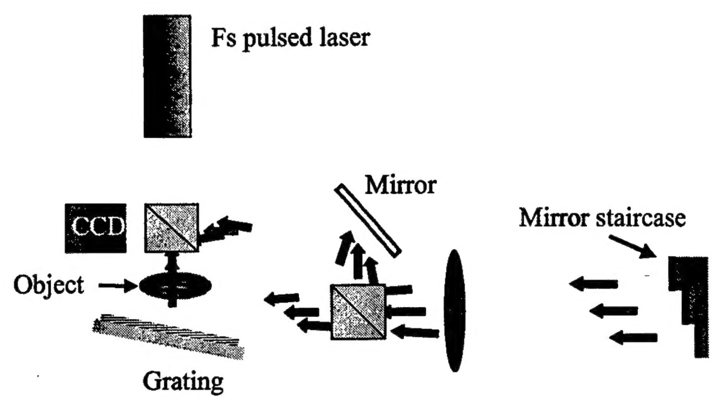

Figure 16. Recording three holograms.

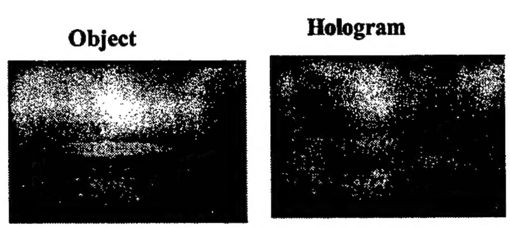

Figure 17. Image of the transparency(left) and hologram recorded on the CCD(right).

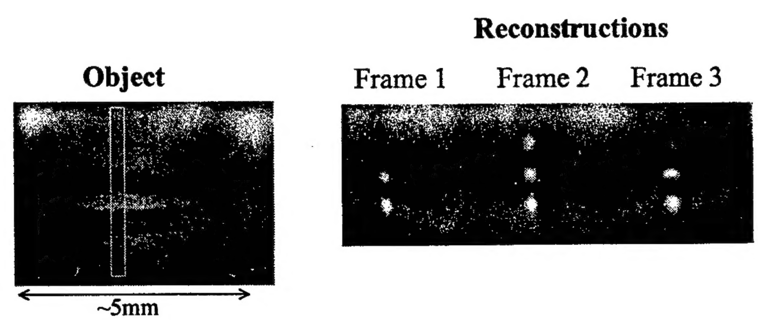

Figure 18. Only a slice of the object is reconstructed.

Publications/Conference Presentations

- "Spectral hole burning in naphthalocyanines derivatives in the region 800 nm for holographic storage applications,", Turukhin AV, Gorokhovsky AA, Moser C, J LUMIN 86 (3-4): 399-405 April, 2000
- "Multiplexing and M# measurement in spectral hole burning medium," CLEO 2001 Technical Digest, pp.374-375.
- "Holographic recording of fast events on a CCD camera", with Zhiwen Liu, Martin Centurion, George Panotopoulos, John Hong and Demetri Psaltis, Optics Letters, Vol. 27, No 1, pp 22-24, January 2002.
- "Holographic recording of fast phenomena", with Zhiwen Liu, Gregory Steckman and Demetri Psaltis, Applied Physics Letters, Vol. 80, No. 5, pp 731-733, February 2002.
- "Derivation and Measurement of M/# for Spectral Hole Burning", *Invited Talk*, SPIE Santa Clara, CA, March, 2002.
- "Holographic Information Systems", SPIE OC 2002, Taiwan ROC, April 2002.
- "Derivation and Measurement of M/# for Spectral Hole Burning", *Invited Talk*, Aerosense, Orlando Fl, April 2002.
- "Multiplexing and M# measurement in spectral hole burning medium," to appear in Journal of Applied Physics